

# Document made available under the Patent Cooperation Treaty (PCT)

International application number: PCT/US05/013327

International filing date: 19 April 2005 (19.04.2005)

Document type: Certified copy of priority document

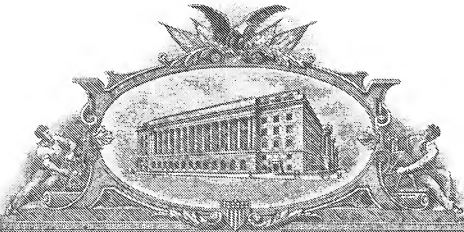
Document details: Country/Office: US  
Number: 60/563,663  
Filing date: 20 April 2004 (20.04.2004)

Date of receipt at the International Bureau: 30 May 2005 (30.05.2005)

Remark: Priority document submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b)



World Intellectual Property Organization (WIPO) - Geneva, Switzerland  
Organisation Mondiale de la Propriété Intellectuelle (OMPI) - Genève, Suisse



# THE UNITED STATES OF AMERICA

TO ALL TO WHOM THESE PRESENTS SHALL COME:

UNITED STATES DEPARTMENT OF COMMERCE

United States Patent and Trademark Office

May 17, 2005

THIS IS TO CERTIFY THAT ANNEXED HERETO IS A TRUE COPY FROM THE RECORDS OF THE UNITED STATES PATENT AND TRADEMARK OFFICE OF THOSE PAPERS OF THE BELOW IDENTIFIED PATENT APPLICATION THAT MET THE REQUIREMENTS TO BE GRANTED A FILING DATE.

APPLICATION NUMBER: 60/563,663

FILING DATE: April 20, 2004

RELATED PCT APPLICATION NUMBER: PCT/US05/13327



Certified by

Under Secretary of Commerce  
for Intellectual Property  
and Director of the United States  
Patent and Trademark Office

Please type a plus sign (+) inside this box ☐

Approved for use through 04/30/2003. OMB 0651-0032  
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

# **PROVISIONAL APPLICATION FOR PATENT COVER SHEET** This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

INVENTOR(S)				
Given Name (first and middle (if any))		Family Name or Surname		Residence (City and either State or Foreign Country)
Shaow Burn		Lin		Midland, Michigan
<input type="checkbox"/> Additional inventors are being named on the _____ separately numbered sheets attached hereto				
TITLE OF THE INVENTION (280 characters max)				
SILICONE VESICLES				
Direct all correspondence to: CORRESPONDENCE ADDRESS				
<input checked="" type="checkbox"/> Customer Number <input type="text" value="00137"/>		<div style="border: 1px solid black; padding: 5px; text-align: center;">             Place Customer Number Bar Code Label here           </div>		
OR Type Customer Number here				
<input type="checkbox"/> Firm or Individual Name				
Address				
Address				
City		State	ZIP	
Country		Telephone	Fax	
ENCLOSED APPLICATION PARTS (check all that apply)				
<input checked="" type="checkbox"/> Specification Number of Pages <input type="text" value="22"/>		<input type="checkbox"/> CD(s), Number <input type="text"/>		
<input type="checkbox"/> Drawing(s) Number of Sheets <input type="text"/>		<input checked="" type="checkbox"/> Other (specify) <div style="border: 1px solid black; padding: 2px;">             Claims - 2 pages Abstract - 1 page           </div>		
<input type="checkbox"/> Application Data Sheet. See 37 CFR 1.76				
METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT (check one)				
<input type="checkbox"/> A check or money order is enclosed to cover the filing fees		FILING FEE AMOUNT (\$)		
<input checked="" type="checkbox"/> The Director is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number <input type="text" value="04-1520"/>		<div style="border: 1px solid black; padding: 5px; text-align: center;"> <b>\$160.00</b> </div>		
<input type="checkbox"/> Payment by credit card. Form PTO-2038 is attached.				
The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.				
<input checked="" type="checkbox"/> No.				
<input type="checkbox"/> Yes, the name of the U.S. Government agency and the Government contract number are: _____				

Respectfully submitted,

SIGNATURE

*Alan Zombek*

TYPED OR PRINTED NAME

Alan Zombek

TELEPHONE

989-496-3101

Date

4-20-04

REGISTRATION NO.

45,260

(if appropriate)

Docket Number:

DC10049PSP1

## **USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT**

This collection of information is required by 37 CFR 1.51. The information is used by the public to file (and by the PTO to process) a provisional application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 8 hours to complete, including gathering, preparing, and submitting the complete provisional application to the PTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Mail Stop Provisional Application, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

P19LARGE/REV05

**UNITED STATES PATENT APPLICATION  
FOR**

5

Silicone Vesicles

10

Having the inventor(s):

Shaow Burn Lin

15

20

**FIELD OF THE INVENTION**

25       The present invention provides a process for preparing a vesicle composition based on mixing an organopolysiloxane having at least one hydrophilic substituent group, a water miscible volatile solvent, and water. The invention also relates to the vesicle compositions produced by the method and their use in various personal, household, and health care applications.

## BACKGROUND OF THE INVENTION

- 5       Silicone surfactants have been designed for various applications by combining a hydrophobic organopolysiloxane with various hydrophilic moieties. For example, the silicone surfactants known as silicone polyethers are based on copolymer structures of polydimethylsiloxane having pendant polyoxyalkylene groups. Such materials find wide utility in many personal, household, and health care compositions as emulsifiers, wetting agents, and general-purpose aqueous surfactants.
- 10       The aggregation behavior of silicone polyethers has been discussed along with their ability to form vesicles. For example, US Patents 5,364,633 and 5,411,744 by Hill teaches the self-assembly of silicone vesicles in aqueous dispersions of certain silicone polyethers. PCT application US03/38455 by Lin teaches the entrapment of various oils in silicone vesicles and their use in various personal care formulations.
- 15       While these references represent advancements in the art, a need still exists to create silicone vesicles having improved stability. For example, the silicone vesicles formed from the self-assembly procedures can lack durability. This lack of durability can limit the processing conditions they can be subject to in the formation of various finished products. Furthermore, such self-assembled silicone vesicles can lack the ability to provide sustained
- 20       release of loaded actives over an extended period of time. Thus, a need exists for a process to prepare silicone vesicles having improved durability and the ability to release loaded actives over an extended period of time.
- The present inventors have discovered a process to prepare silicone vesicles that provides such advantages.

### SUMMARY OF THE INVENTION

5       The present invention provides a process for preparing a vesicle composition comprising;

I) mixing,

A) an organopolysiloxane having at least one hydrophilic substituent group,

B) a water miscible volatile solvent,

10       C) water,

to form an aqueous dispersion of the organopolysiloxane,

II) removing the water miscible volatile solvent from the aqueous dispersion to form the vesicle composition.

15

The present invention also relates to the vesicle compositions obtained from the inventive process, the vesicle compositions further comprising a personal, household, health care ingredient.

20       The present invention further relates to personal, household, health care product compositions containing the vesicle compositions.

## DETAILED DESCRIPTION OF THE INVENTION

The first step in the process of the present invention comprises mixing;

- 5           A) an organopolysiloxane having at least one hydrophilic substituent group,  
          B) a water miscible volatile solvent,  
          C) water,

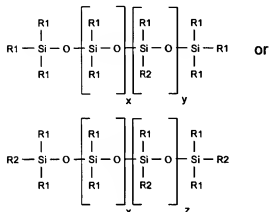
to form an aqueous dispersion of the organopolysiloxane.

10           Component A) is an organopolysiloxane having at least one hydrophilic substituent group. Organopolysiloxanes are well known in the art and are often designated as comprising any number of "M" siloxy units ( $R_3SiO_{0.5}$ ), "D" siloxy units ( $R_2SiO$ ), "T" siloxy units ( $RSiO_{1.5}$ ), or "Q" siloxy units ( $SiO_2$ ) where R is independently any hydrocarbon group. In the present invention, the organopolysiloxane has at least hydrophilic substituent. That is, at least one of the R hydrocarbon groups present in the organopolysiloxane is a hydrophilic group. For purposes of  
15 this invention, "hydrophilic group" is the accepted meaning in the art, i.e., designating water loving chemical moieties. Thus, the hydrophilic group can be selected from various cationic, anionic, zwitterionic, polyoxyalkylene, oxoazoline chemical moieties that are commonly used in combination with various hydrophobic chemical moieties to create surfactant structures or molecules having surface-active behavior.

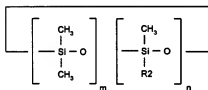
20           The amount of the hydrophilic substituent on the organopolysiloxane can vary, depending on the specific chemical component, providing there is at least one hydrophilic group present on the organopolysiloxane. However, the amount of the hydrophilic groups present in the organopolysiloxane can be described by its weight percent, or in particular, the weight percent of the organopolysiloxane and weight percent of the total hydrophilic groups present in  
25 the molecule. Typically, the weight percent of the siloxane units in the organopolysiloxane can vary from 20 to 85, alternatively from 30 to 85, or alternatively from 35 to 80 weight percent, while the remaining weight portion of the organopolysiloxane is the hydrophilic group.

          In one embodiment of the present invention, the organopolysiloxane having at least one hydrophilic substituent group is selected from silicone polyethers. Silicone polyethers  
30 (SPEs) generally refer to silicones containing polyether or polyoxyalkylene groups, which could take in many different structural forms. One such form is rake-type SPEs which are derived most commonly from hydrosilylation SiH sites of  $MD_xD'_yM$  with allyloxy-functional

polyethers in the presence of Pt catalyst. In this embodiment, component (A) is a silicone polyether and can have a structure represented by:



A cyclic polyether of the type shown below can also be used.



5

In these structures, R1 represents an alkyl group containing 1-6 carbon atoms such as methyl, ethyl, propyl, butyl, pentyl, and hexyl; R2 represents the group - (CH<sub>2</sub>)<sub>a</sub>O(C<sub>2</sub>H<sub>4</sub>O)<sub>b</sub>(C<sub>3</sub>H<sub>6</sub>O)<sub>c</sub>R<sub>3</sub>; x has a value of 1-1,000, alternatively 1 - 500, or alternatively 10 - 250; y has a value of 1-500, alternatively 1 - 100, or alternatively 2 - 50; z has a value of 1-500, or alternatively 1 - 100; m has a value of 3-5; n is one; a has a value of 3-6; b has a value of 4-20; c has a value of 0-5; and R<sub>3</sub> is hydrogen, a methyl group, or an acyl group such as acetyl. Typically, R1 is methyl; b is 6-12; c is zero; and R<sub>3</sub> is hydrogen.

The silicone surfactants can be prepared by any of the techniques known in the art, and many are commercially available.

15

The process of the present invention is particularly useful to prepare silicone vesicle compositions containing a silicone polyether where the silicone polyether has a D/D' ratio ranging from 5/1 to 50/1, alternatively from 10/1 to 50/1 or alternatively from 20/1 to 50/1.



Component B) is a water-miscible volatile solvent. As used herein "water-miscible" means the solvent forms a dispersion with water at room temperature for at least several hours. "Volatile" means the solvent has a higher vapor pressure than water at various temperatures. As such, when the aqueous dispersion of the organopolysiloxane and solvent  
5 are subjected to conditions to remove the solvent, such as heating the dispersion under reduced pressures, the solvent is primarily removed first, allowing all or most of the water to remain in the composition.

Suitable water-miscible volatile solvents for vesicle dispersion preparation include organic solvents such as alcohols, ethers, glycols, esters, acids, halogenated hydrocarbons,  
10 diols. The organic solvents should be miscible with water at the proportion and lower in order to effectively disperse silicones and maintain stable and uniform dispersion overtime. For the purpose of illustration, water-miscible alcohols include methanol, ethanol, propanol, isopropanol, butanol, and higher hydrocarbon alcohols; ethers include glycol ethers, methyl-ethyl ether, methyl isobutyl ether (MIBK), etc; glycols include propylene glycols, esters  
15 include esters of triglycerol, the esterification products of acid and alcohol; halogenated hydrocarbons include chloroform. Typically water-miscible organic solvents are solvents with relatively low boiling points (<100°C) or high evaporation rate, so they may be removed under vacuum with ease. The most preferred water-miscible organic solvents for this invention are volatile alcohols including methanol, ethanol, isopropanol, and propanol. These  
20 alcohols can be removed from aqueous mixtures containing silicone vesicle dispersions via vacuum stripping at ambient temperature.

The order of mixing components A), B), and C) is not critical, but typically A) and B) are first mixed and then water added to the mixture. There are no special requirements or conditions needed for effecting the mixing of components A), B), and C). The mixing can  
25 be conducted in a batch, semi-continuous, or continuous process.

The amount of components A), B), and C) can vary in the process, but typically range as follows;

- A) 2 to 50 wt%, alternatively 2 to 25 wt %, or alternatively 2 to 15 wt%,
- B) 2 to 50 wt%, alternatively 2 to 25 wt %, or alternatively 2 to 15 wt%,
- 30 C) sufficient amount to provide the sum of the wt% of A), B), and C) to equal 100%

The amount of B) water-miscible volatile solvent used to disperse the organopolysiloxane depends on the type of organopolysiloxane and how much hydrophilic

groups are present. Typically, the aqueous mixture to effectively disperse silicones comprises of 5 to 80 parts of solvent and 20 to 95 parts of water; alternatively 5 to 50 parts of water, or alternatively 10 to 40 parts water.

5 The present invention also relates to the vesicle compositions produced by the methods, as described supra. The formation of vesicles can be confirmed by techniques common in the state of the art. Typically, vesicles having a lamellar phase structure which exhibit birefringence when examined with a cross polarizing microscope. Alternatively, the formation of vesicles can be demonstrated by Cyro-Transmission Electron Microscopy (Cryo-TEM) techniques. Particle size measurements can also be used to indicate that the  
10 organopolysiloxanes are sufficiently dispersed in aqueous medium typical of vesicle sizes. For example, average particle sizes of less than 0.500  $\mu\text{m}$  (micrometers), are typical for dispersed vesicles. Vesicles having a average particle size of less than 0.200  $\mu\text{m}$ , or 0.100  $\mu\text{m}$  are possible with the method of the present invention.

The present invention further relates to vesicle compositions further comprising a  
15 personal, household, or health care ingredient. Thus, the vesicle compositions can be used to entrap, and subsequently deliver after application, a personal, household care, or health care ingredient. A listing of possible personal, household, or health care ingredients is taught in WO 03/101412, which is incorporated herein by reference. The personal or health care ingredient can also be selected from a personal or health care "active", that is, any compound  
20 known to have either cosmetic and/or pharmaceutical activity. A representative listing of such personal or health care actives are disclosed in US Patent 6,168,782, which is hereby incorporated by reference.

Compositions prepared according to the invention can be used in various over-the-counter (OTC) personal care compositions, health care compositions, and household care  
25 compositions, but especially in the personal care arena. Thus, they can be used in antiperspirants, deodorants, skin creams, skin care lotions, moisturizers, facial treatments such as acne or wrinkle removers, personal and facial cleansers, bath oils, perfumes, colognes, sachets, sunscreens, pre-shave and after-shave lotions, liquid soaps, shaving soaps, shaving lathers, hair shampoos, hair conditioners, hair sprays, mousses, permanents,  
30 depilatories, hair cuticle coats, make-ups, color cosmetics, foundations, blushes, lipsticks, lip balms, eyeliners, mascaras, oil removers, color cosmetic removers, nail polishes, and powders.

## EXAMPLES

The following examples are presented to further illustrate the compositions and methods of this invention, but are not to be construed as limiting the invention. All parts and percentages in the examples are on a weight basis and all measurements were obtained at about 23°C, unless indicated to the contrary.

The vesicle compositions were analyzed via Cyro-TEM techniques according to the following procedure. Around 2.3 µl of aqueous sample solution was loaded using a micropipette on a lacey carbon film coated Cu TEM grid that was cleaned and rinsed with acetone and chloroform. The samples were diluted to about 5% solution with de-ionized water. The excess fluid on the grid surface was removed by blotting the surface with a filter paper for 1.5 second to make an aqueous thin film for TEM. The grid was then plunged into a liquid ethane contained in a small vessel located in a larger liquid nitrogen vessel under -175 °C atmosphere in the cryo-plunge system to vitrify the water film on the grid and to avoid water crystallization. The quenched sample grid was transferred in to the cryo-grid box in the cryo-plunge system. The grid box containing the sample was transferred into a Gatan cryo-transfer system filled with liquid nitrogen and loaded in a cryo-TEM stage, which has been positioned in the cryo-transfer system and cooled down to below -160 °C. The sample was loaded in TEM (JEOL 2000FX) and the images were observed at below -160 °C. A much colder finger, cooled to -180 °C in TEM using liquid nitrogen, was present to reduce any possible contamination on the cold specimen surface under high vacuum during TEM analysis. The digital images, as shown herein, were taken using a Gatan CCD camera attached at the bottom of the TEM column and Digital Micrograph software.

**Example 1** (Reference example)*Silicone polyethers used for preparation of vesicle dispersions*

5 A series of silicone polyethers were prepared, as illustrated in Table 1 below. This series were "rake type" silicone polyethers since they were derived from various dimethyl-methylhydrogen-polysiloxanes of the general formula,  $MD_xD'_yM$  and allyloxy polyethers. The  $MD_xD'_yM$  siloxanes were prepared according to known techniques. The silicone polyethers were prepared using well know platinum catalyzed hydrosilylation techniques, such as those disclosed in US Patent 2,823,218.

The D/D' ratio corresponds to x / y ratio in the formula  $MD_xD'_yM$  and total DP is the sum of x and y. EO7 is allyloxy poly(ethylene oxide) having about seven EO repeat units, and EO12 is allyloxy poly(ethylene oxide) having about 12 EO repeat units; both were obtained from Dow Chemical Company.

15

**Table 1**

Siloxane structure	$MD_{27}D'_{31}M$	$MD_{12}D'_{12}M$	$MD_{28.8}D'_{1.12}M$	$MD_{46.88}D'_{3.12}M$	$MD_{70}D'_{31}M$
D / D' ratio	9.0	10	15	15	23.3
Total dp, D+D'	30	24	30	50	73
EO7 polyether		SPE 1	SPE 2	SPE 3	
EO12 polyether	SPE 4	SPE 5	SPE 6	SPE 7	SPE 8

For  $MD_xD'_yM$  rake-type SPEs, D/D' ratio represents the relative ratio of the number of siloxane repeat units to the number of polyether substituted units on the siloxane polyethers, and a direct measure of hydrophobic to hydrophilic ratio of the SPEs. The lower the D/D' ratio, the more hydrophilic the SPEs, and more readily dispersible the SPE is in water.

20

**Example 2***Effect of D/D' ratio of SPEs and alcohol on vesicle dispersion preparation*

5 SPEs were generally found to spontaneously disperse in water, forming vesicles, when the D/D' was 12 or lower, as summarized in Table 2 below. When the D/D' ratio of the SPE was 15 or higher, the SPEs were less dispersible in water, and the rate of SPE dispersion in water could be very slow to incomplete. This example demonstrates the time needed to disperse SPE 6 (from Example 1) in water and how much faster it disperses when a small amount of isopropanol was present.

10 When D/D' was greater than 15, the SPEs did not spontaneously disperse in water. For example, the SPEs derived from MD70D'3M (such as SPE 8) were not dispersible at all in water, even over more than a period of 2 months.

Examples 2A and 2B show how SPE 6, a silicone polyether of MD<sub>28.8</sub>D'<sub>1.22</sub>M siloxane structure dispersed in water alone and in the presence of 5 % isopropanol, respectively. It  
15 took about 3 weeks of continuous mixing to form a homogeneous dispersion in water as opposed to less than 1 day when 5 % by weight of isopropanol present in water.

Homogeneous dispersions of SPE 3 and SPE 7, silicone polyethers of MD<sub>46.88</sub>D'<sub>3.12</sub>M siloxane backbone with D/D' of 15, were also made into homogeneous dispersions within 2 days with aid of 5 % isopropanol, as shown in Examples 2C and 2D.

Table 2

Example	2A	2B	2C	2D
Process History	3 weeks of continuous mix	As mixed, uniform dispersion in 1 day	As mixed, homogeneous in 2 days	As mixed, uniform dispersion in 2 days
Formulation				
SPE 3 (D/D' = 15), g			10	
SPE 6 (D/D' = 15), g	10.02	10.09		
SPE 7, (D/D' = 15) g				10.03
Isopropanol, g		5.02	5.000	5.02
D.I. Water, g	90.030	85.3	85.000	85.3
Final Dispersion Composition				
Wt. % SPE	10.00	10.00	10.00	10.00
Wt. % Isopropanol	0.00	5.00	5.00	5.00
Wt. % Water	90.00	85.00	85.00	85.00
pH of the dispersion	5.99	6.52	6.2	6.22
Avg. dispersion size, um				
D(v, 0.5), um				
D(v, 0.9), um				

- 5 Cryo-TEM images of the vesicles formed in Examples 2A, 2B, and 2D are shown in figures 1 – 3 respectively. These images show the presence of single lamellar vesicles in Example 2A, and presence of multi-lamellar vesicles in Examples 2B and 2D.

**Example 3***Comparative example*

- 5           A dispersion of SH 3775M SPE in water was attempted by incorporating 10.0 g of SH 3775M SPE into 90.0 g of de-ionized water in a glass jar. The jar was subjected to mixing on a rotowheel mixer. Even after 4 weeks of mixing the SPE remained as separated in water, no dispersion was formed.

10       **Example 4**

*Preparation of 20% and 30% SPE 8 Vesicle Dispersions*

*Example A4: Vesicle dispersion of 14.4% SPE 8 dispersion in 28.7% isopropanol and 56.9 % water.*

- 15           A dispersion was prepared by first adding SPE 8 into isopropanol with continuous mixing. Then, water was gradually added with continuous stirring. The resulting final mixture was clear, and a homogenous dispersion. SPE vesicles of 31 nm average sizes were observed and the dispersion has a clear water-like appearance due to the size of the vesicles.

- 20       *Example 4B: Vesicle dispersion of 20.0% SPE 8 in 80.0 % water.*

- A homogeneous dispersion of SPE 8 in isopropanol and water was prepared, following a similar procedure for Example 4A. This dispersion was further stripped under vacuum at ambient temperature to remove volatile isopropanol using a Rotovapor. The theoretical amount of 74.93 g of IPA was completely removed and yielded a theoretically alcohol-free aqueous dispersion of SPE 8 vesicles. The resultant product was a milky-white, homogeneous dispersion with an average particle size of 212 nm, as measured by Nanotrac particle analyzer.

- 30       *Example 4C: vesicle dispersion of 21.2% SPE 8 in 28.8% isopropanol and 50% water.*

- A homogeneous vesicular dispersion of 21 nm average particle size in isopropanol and water was prepared, following a procedure similar to that of Example 4A.

*Example 4D: vesicle dispersion of 29.5 % SPE 8 in 1.0% isopropanol and 69.5% water*

- A homogeneous vesicular dispersion of 436 nm average particle size in water was prepared, following a procedure similar to that of Example 4B. The final mixture was a milky-white, homogenous dispersion.

Example #	4A	4B	4C	4D
Process History	As mixed	Mixed, then stripped	As mixed	Mixed, then stripped
SH 3775M SPE, g	43.11	37.59	64.12	55.89
IPA, g	86.23	74.93	87.00	75.93
D.I. Water, g	170.724	148.55	151.268	131.82
Batch size, as mixed	300	261.07	302.4	263.64
IPA Removed, g		-75.70		-74.10
Final batch, after strip		185.37		189.53
Wt. % Polymer	14.4	20.0	21.2	29.5
Wt. % IPA	28.7	0.0	28.8	1.0
Wt. % Water	56.9	80.0	50.0	69.5
Appearance of dispersion	Clear water-like liquid	Milky white material	Clear water-like liquid	Milky white material
Avg. particle size, $\mu\text{m}$	0.031	0.212	0.021	0.436
D(v, 0.5), $\mu\text{m}$	0.029	0.178	0.019	0.353
D(v, 0.9), $\mu\text{m}$	0.044	0.428	0.036	0.914



**Example 5*****Effect of isopropanol concentration on vesicle dispersion size***

5       The ease of dispersing silicone polyethers and, in some cases, the size and structure of  
the vesicles in the final water dispersions depends on the amount of isopropanol. This  
example illustrates this effect with two rake-type SPEs, SPE 7 and 8 as identified in  
Example 1.

***Examples 5A, 5B, and 5C***

10

Dispersions of SPE 8 in water containing 5% to 20 wt.% isopropanol were prepared  
according to the procedure described previously, according to the formulations summarized  
in Table 5. Example 5C was prepared by subjecting the Example 5B dispersion to further  
vacuum stripping to remove most of the IPA.

15

***Examples 5D and 5E***

A homogeneous vesicle dispersion of SPE 7 was made in 20 % IPA and 70 % water  
having an average dispersion size of only 77 nm, according to the previously described  
procedure, as summarized as Example 5D. The isopropanol was removed by vacuum  
20 stripping, resulting in a dispersion (Example 5E) having an average dispersion size of only 49  
nm in water, as summarized in Table 5.

Table 5

Example #	5A	5B	5C	5D	5E
Process History	As mixed	As mixed	Mixed, then stripped	As mixed	Mixed, then stripped
SH 3775M SPE, g	10.025	30.012	20		
18841-91 SPE, g				30.006	20.0
IPA, g	10.005	60.082	10	60.172	40.0
D.I. Water, g	80.044	210.063	140	210.757	140.0
Final Dispersion Composition					(-40 g alcohol)
Wt. % Polymer	10.02	10.00	11.76	9.97	12.50
Wt. % IPA	10.00	20.02	5.88	20.00	0.00
Wt. % Water	79.98	69.98	82.35	70.03	87.50
pH of the dispersion	5.87	7.14	6.81	6.35	6.38
Avg. dispersion size, um	1.427	0.106	0.099	0.077	0.049
D(v, 0.5), um	0.955	0.092	0.091	0.071	0.072
D(v, 0.9), um	3.789	0.178	0.156	0.112	0.104

- 5 Cryo-TEM images confirmed the silicone polyether dispersions were nano-scale vesicles and aggregates of vesicles. The actual sizes observed were consistent with the particle size measured by Nanotracs particle analyzer. The cryo-TEM images of SPE 8 dispersion (Example 5C) and that of SPE 7 dispersion (Example 5E) are shown in figures 4 and 5 respectively.

10

### Example 6

*SPE vesicle dispersions containing Vitamin A palmitate*

- 15 The method of this invention can also be used to entrap and retain oil-soluble substances into vesicles in aqueous dispersions. This is very desirable and beneficial utility of vesicles and vesicular aggregates prepared from silicone polyethers. As many personal care actives & ingredients are not water-soluble or not water-compatible. This makes it not practical to incorporate such ingredients as vitamins, fragrances, sunscreens, and oils directly

into water-based or water-continuous end-product formulations, such as creams, lotions, gels, etc.

This series of examples shows that vitamin A palmitate (a preferred form of vitamin A for skin care products) can be successfully incorporated into SPE based vesicles in water  
5 dispersions, using the method of this invention. Results and formulations are summarized in Table 6.

#### *Examples 6A and 6B*

*Vitamin A palmitate entrapped SPE vesicles in partially and fully stripped aqueous  
10 dispersions.*

Vitamin A palmitate (VAP) was first dispersed in isopropanol at 50/50 ratio by weight to keep VAP in a liquid form for ease of mixing. The VAP/IPA was then mixed in with SPE 8, followed by incorporation of isopropanol to form a homogeneous mixture. While under continuous mixing, water was gradually and continuously incorporated into the mixture  
15 containing the SPE, VAP and IPA. Then, the mixture was homogenized, using an APV benchtop homogenizer, to further narrow and reduce the dispersion size. The homogenized dispersion was subsequently stripped to remove isopropanol under vacuum at ambient temperature, using a Rotovapor.

The 6A example was a partially stripped dispersion having about 10% IPA remaining  
20 and an average dispersion size of 0.229  $\mu\text{m}$ . The 6B example was a fully stripped dispersion containing 20.8% SPE, 4.5% VAP, 2.2% IPA and 72.5% water. Example 6B was a homogeneous, beige milky dispersion having an average vesicle size of 0.974  $\mu\text{m}$ .

#### *Example 6C*

*Vitamin A palmitate loaded dispersion made without homogenization using ethano.*

This dispersion was prepared by first dispersing vitamin A palmitate into SPE 8 in the presence of a small quantity of ethanol. The prescribed amount of ethanol was mixed in with SPE / VAP to form a homogeneous mixture. While under continuous mixing, water was  
30 gradually and continuously incorporated into the mixture. A homogeneous mixture was obtained. The mixture was then subjected to vacuum stripping to remove the volatile alcohol.

The final dispersion had a composition of 4.5% VAP, 20.7% SPE, 2.9% EtOH and 72.0 % water. The dispersion had an average dispersion size of 0.763  $\mu\text{m}$ .

*Examples 6D and 6E*

5 *Vitamin A palmitate loaded dispersions made with EtOH.*

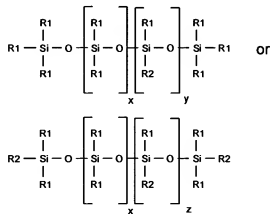
Ethanol was used as dispersing aid to prepare these VAP loaded SPE vesicles in water dispersions. The procedures followed the ones that used to make Example 6A and 6B, except EtOH used in place of IPA. The final dispersion compositions are shown in Table 6. The two dispersions had an average dispersion size of 0.284  $\mu\text{m}$  and 0.331  $\mu\text{m}$ , respectively.

**Table 6**

Example #	6A	6B	6C	6D	6E
Alcohol type used	IPA	IPA	EtOH	EtOH	EtOH
Process History	Mixed, Homog., Partially Stripped	Mixed, Homog., Fully Stripped	Mixed, Stripped	Mixed, Homog., Partially Stripped	Mixed, Homog., Fully Stripped
VAP (vitamin A palmitate), g	3.96	3.91	9.89	3.94	3.69
Alcohol, g	32.71	32.32	81.61	32.54	30.50
SPE 8, g	18.35	18.13	45.73	18.23	17.09
Water, g	63.96	63.19	159.01	63.40	59.42
Batch size, as mixed, g	118.97	117.55	296.23	118.11	110.70
Alcohol Removed, g	23.48	30.37	75.30	22.21	28.47
Batch size, after strip, g	95.49	87.18	220.93	95.90	82.23
Final Dispersion composition					
Wt. % VAP	4.1	4.5	4.5	4.1	4.5
Wt. % Alcohol	9.7	2.2	2.9	10.8	2.5
Wt. % SPE Polymer	19.2	20.8	20.7	19.0	20.8
Wt. % Water	67.0	72.5	72.0	66.1	72.3
Appearance of Dispersion	Beige milky dispersion	Beige milky dispersion	Beige milky dispersion; small oil droplets atop	Beige milky dispersion	Beige milky dispersion
Average particle size, $\mu\text{m}$	0.2291	0.974	0.763	0.2842	0.331
D(v, 0.5), $\mu\text{m}$	0.1602	0.995	0.374	0.1624	0.2103
D(v, 0.9), $\mu\text{m}$	0.513	1.783	1.987	0.727	0.785

## CLAIMS

1. A process for preparing a vesicle composition comprising;
- 5        I) mixing,
- A) an organopolysiloxane having at least one hydrophilic substituent group,
- B) a water miscible volatile solvent, and
- C) water,
- to form an aqueous dispersion of the organopolysiloxane,
- 10       II) removing the water miscible volatile solvent from the aqueous dispersion
- to form the vesicle composition.
2. The process of claim 1 wherein the aqueous dispersion contains;
- A) 5 to 50 wt % of the organopolysiloxane,
- 15       B) 5 to 50 wt% of the water miscible solvent,
- C) sufficient amount of water
- to provide the sum of the wt% of A), B), and C) to equal 100 wt %.
3. The process of claim 1 wherein the organopolysiloxane is a silicone polyether.
- 20       4. The process of claim 1 wherein the organopolysiloxane is a silicone polyether having a structure represented by:



where R1 represents an alkyl group containing 1-6 carbon atoms;

R2 represents the group  $-(CH_2)_aO(C_2H_4O)_b(C_3H_6O)_cR_3$ ;

x is 1-1,000; y is 1-500; z is 1-500; a is 3-6; b is 4-20; c is 0-5;

5 and R3 is hydrogen, a methyl group, or an acyl group.

4. The process of claim 1 wherein the water miscible volatile solvent is an alcohol.

5. The process of claim 1 wherein the water miscible volatile solvent is isopropyl alcohol.

10

6. The product resulting from the process of any one of claims 1 – 5.

7. A personal, household, or health care product comprising the product of claim 6.

15

**ABSTRACT**

5           A process for preparing a vesicle composition based upon mixing an organopolysiloxane having at least one hydrophilic substituent group, a water miscible volatile solvent, and water is disclosed. The vesicle compositions produced by the method are useful in various personal, household, and health care applications.

10

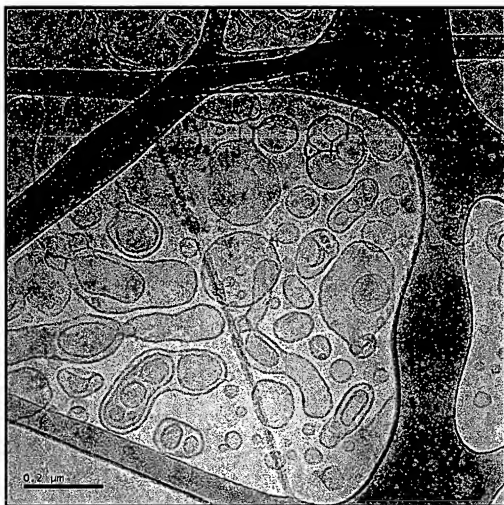
15

20

25

30

Figure 1



BEST AVAILABLE COPY



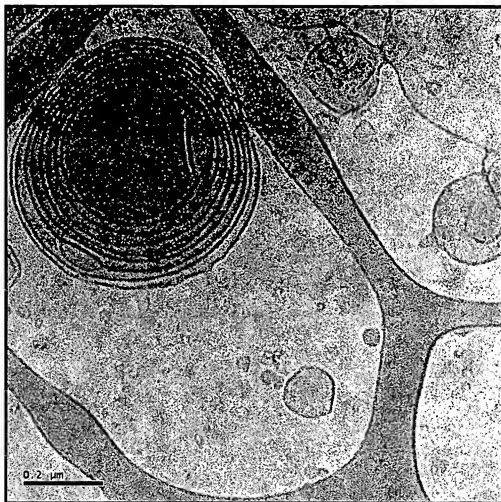
Figure 2



BEST AVAILABLE COPY

Figure 3

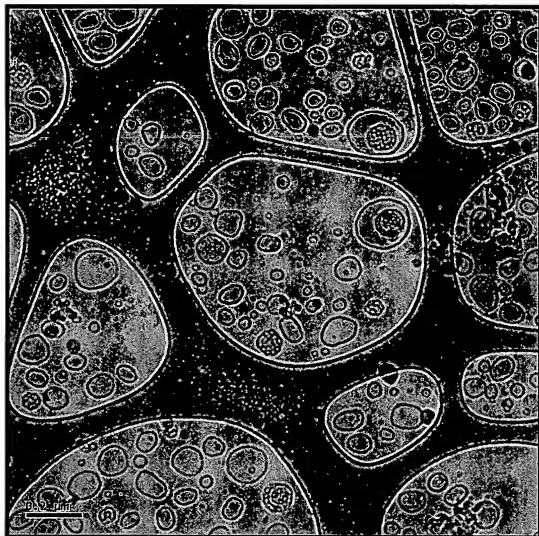
5



BEST AVAILABLE COPY

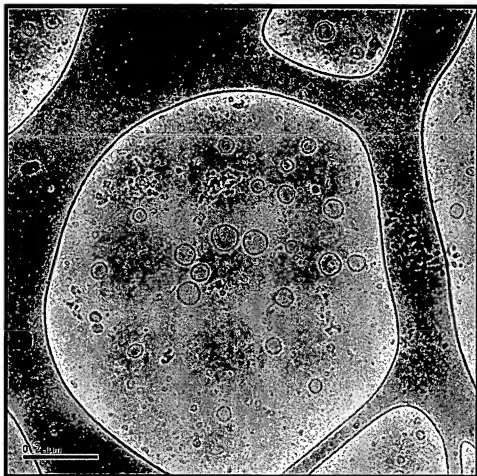
Figure 4

5



BEST AVAILABLE COPY

Figure 5



BEST AVAILABLE COPY